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Review

Application of Rejuvenators in Asphalt Binders: Classification and Micro- and Macro-Properties

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Abstract

Rejuvenating aged asphalt is critical for sustainable road construction and resource utilization. This paper systematically reviews the current research on rejuvenators, focusing on their classification and the micro-, and macro-properties of rejuvenated asphalt. Rejuvenators are categorized into mineral oil-based, bio-based, and compound types. Each type offers distinct advantages in recovering the performance of aged asphalt. Mineral oil-based rejuvenators primarily enhance low-temperature cracking resistance through physical dilution, while bio-based rejuvenators demonstrate superior environmental sustainability and stability. Compound rejuvenators, particularly those incorporating reactive compounds, show the best results in repairing degraded polymer modifiers and improving both low- and high-temperature properties of aged, modified asphalt. Atomic Force Microscopy (AFM), Fluorescence Microscopy (FM), and Scanning Electron Microscopy (SEM) have been applied to analyze the micro-properties of rejuvenated asphalt. These techniques have revealed that rejuvenators can restore the microstructure of aged asphalt by dispersing agglomerated asphaltenes and promoting molecular mobility. Functional groups and molecular weight changes, characterized by Fourier Transform Infrared Spectroscopy (FTIR) and Gel Permeation Chromatography (GPC), indicate that rejuvenators effectively reduce oxidation products and molecular weight of aged asphalt, restoring its physicochemical properties. Macro-property evaluations show that rejuvenators significantly improve penetration, ductility, and fatigue resistance. Finally, this review identifies the key characteristics and challenges associated with rejuvenator applications and provides an outlook on future research directions.

Keywords: road engineering; rejuvenator; reclaimed asphalt pavement; rejuvenation effect

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1. Introduction

Asphalt mixture gains extensive application within road paving projects, with global annual consumption exceeding 1 billion tons [1,2]. As asphalt pavements age, road rehabilitation projects generate substantial amounts of RAP. If not adequately managed, RAP disposal can contribute to environmental pollution and result in significant resource wastage [3–6]. With the growing global focus on sustainable resource utilization and the decrease in crude oil storage, the development of efficient RAP rejuvenation technologies has become a critical research priority.

Current research primarily explores the reuse of RAP by incorporating rejuvenators to restore the properties of aged asphalt [7,8]. When virgin asphalt ages, its colloidal properties

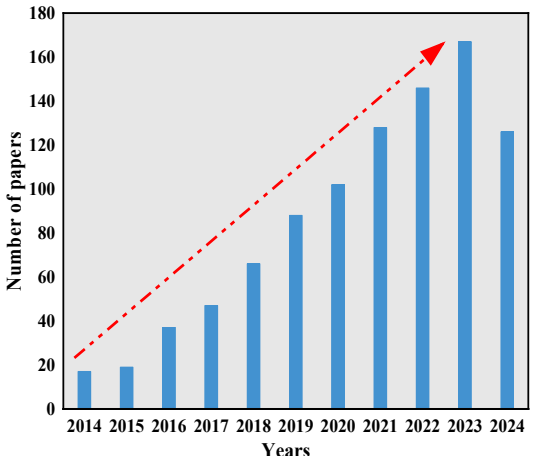
may change; such changes include a reduction in light components, the conversion of maltenes into asphaltenes, and the oxidation of other constituents [9]. In contrast, the aging process of modified asphalt is more complicated. It covers not only the deterioration of the virgin asphalt but also the oxidation and degradation of polymer additives [10,11]. These fundamental differences underscore the necessity of selecting appropriate rejuvenators and developing tailored performance recovery strategies for different types of asphalt.

In the case of virgin asphalt, the primary function of rejuvenators is to rejuvenate the phase equilibrium between the discrete and continuous phases. Rejuvenators can improve molecular mobility by reducing asphaltenes content and enhancing phase dispersion [5,12]. These processes decrease the viscosity of aged virgin asphalt, ultimately restoring its properties [13–15]. In the case of modified asphalt, rejuvenators can not only recover the properties of the virgin asphalt but also repair or supplement the oxidized or degraded polymer modifier, further enhancing the properties of aged modified asphalt [16]. Rejuvenators are typically categorized into mineral oil-based, bio-based, and compound types.

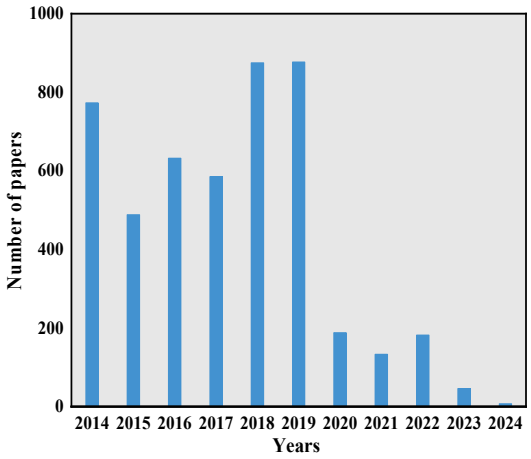
Earlier, Shen et al. [17] restored the workability of aged asphalt using a soft binder. Building on this, Cao et al. [18] found that incorporating a rejuvenator into asphalt, rather than using virgin asphalt, resulted in a more uniform rejuvenated asphalt mixture system. Subsequently, Asli et al. [19] were the first to apply WCO for the regeneration of aged asphalt, significantly restoring its properties. Xu et al. [20] researched how a mineral oil-based rejuvenator affected aged virgin asphalt. They discovered that the rejuvenator enhanced low-temperature cracking resistance and increased service life. Suo et al. [21] compared the effectiveness of mineral oil-based and bio-based rejuvenators, reporting that bio-based rejuvenators resulted in lower weight loss during aging, along with improved stability and superior properties. Additionally, to address the aging of polymer-modified asphalt, Ma et al. [22] restored part of aged polymer-modified asphalt's performance using a solid modifying additive. Hong et al. [23] developed a compound rejuvenator combining AO and SBS, which could enhance the cracking resistance of rejuvenated asphalt at low temperatures and mitigate excessive softening caused by AO. Furthermore, research teams led by Xu et al. [24] and Cao et al. [25] found that certain compounds containing epoxy and isocyanate functional groups can repair degraded SBS modifiers. In addition, Xu et al. [24] and Xing et al. [16] conducted detailed reviews of current reactive rejuvenation systems, but no one has conducted a detailed evaluation of the differences between light oil rejuvenation systems and the reactive rejuvenation systems. When used in combination with light oils, these compounds significantly restore the properties of aged SBSMA. These findings indicate that different rejuvenators exhibit varying rejuvenation effects, necessitating further in-depth research to optimize their selection and application.

Bibliometric analysis reveals that research on asphalt rejuvenators has garnered significant attention in recent years. A search on Web of Science using "asphalt" and "rejuvenator" as keywords retrieves 1142 relevant papers. Notably, all of these papers were published within the last two decades, highlighting the growing interest in rejuvenator research. Statistical data further indicates that 1103 of these papers were published in the past decade (since 2014), with a steady annual increase in publication volume, as illustrated in Figure 1a. This trend underscores the emergence of rejuvenators as a prominent research focus within the scientific community. As depicted in Figure 1b, papers published between 2014 and 2019 have been cited frequently, reflecting significant advancements and key milestones in the field. The relatively lower citation frequency of papers published after 2020 is expected, as these studies are still recent. Additionally, Figure 1c shows the average citation rate of publications over the years, with a peak between 2014 and 2019, providing a foundation that supports future research and advances and further promoting the development of rejuvenator technologies. Figure 1d demonstrates China and the United States' status as

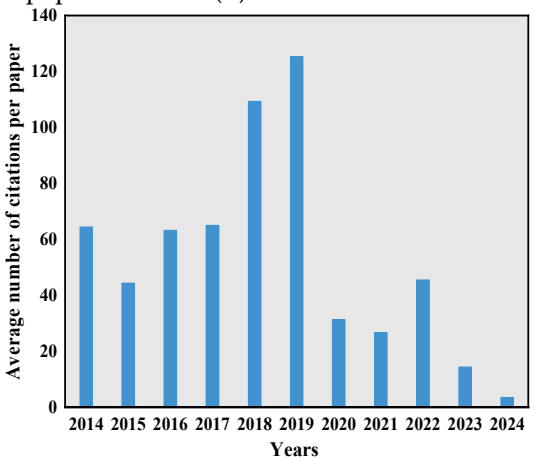
leaders in both number of publications and citation frequency, underscoring their substantial contributions to rejuvenator research and their dominant role in advancing global knowledge in this area.



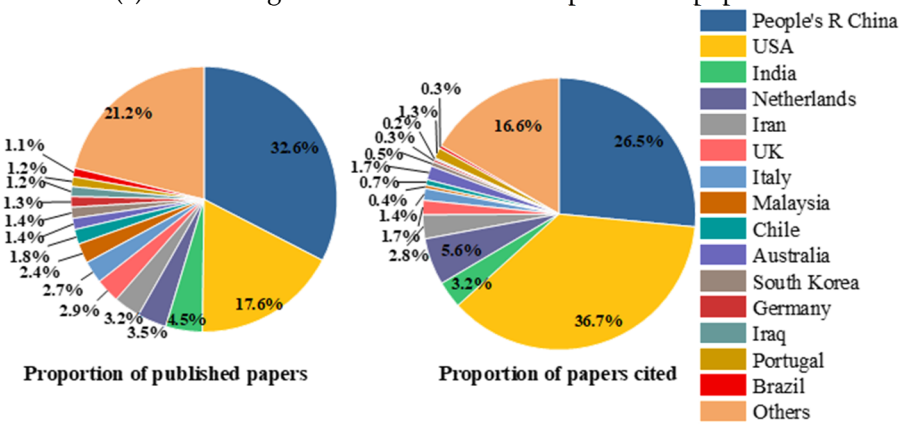
(**a**) The number of published papers.



(**b**) The cumulative number of citations of published papers.



(**c**) The average number of citations of published papers.



(**d**) Papers published in each country and the proportion of papers cited.

Figure 1. Analysis of research results in recent years: (**a**) The number of published papers, (**b**) The cumulative number of citations of published papers, (**c**) The average number of citations of published papers (**d**) Papers published in each country and the proportion of papers cited.

Although extensive research has been conducted on individual types of rejuvenators, comparative studies evaluating the three main categories of rejuvenators remain limited. To address this gap, this paper comprehensively reviews the current status of different rejuvenator types in asphalt rejuvenation. First, the paper introduces the classification of

rejuvenators. Then, it systematically examines the impact of multifarious rejuvenators on aged asphalt, analyzing both micro- and macro-property changes. Finally, it discusses the challenges associated with rejuvenator application and outlines future research directions in this field.

2. Classification of Rejuvenators

Rejuvenators are specialized additives designed to restore aged asphalt's physicochemical and bonding properties [26–29]. Based on existing studies [24,30–32], rejuvenators can be categorized into three categories: mineral oil-based rejuvenators, bio-based rejuvenators, and compound rejuvenators, as shown in Figure 2.

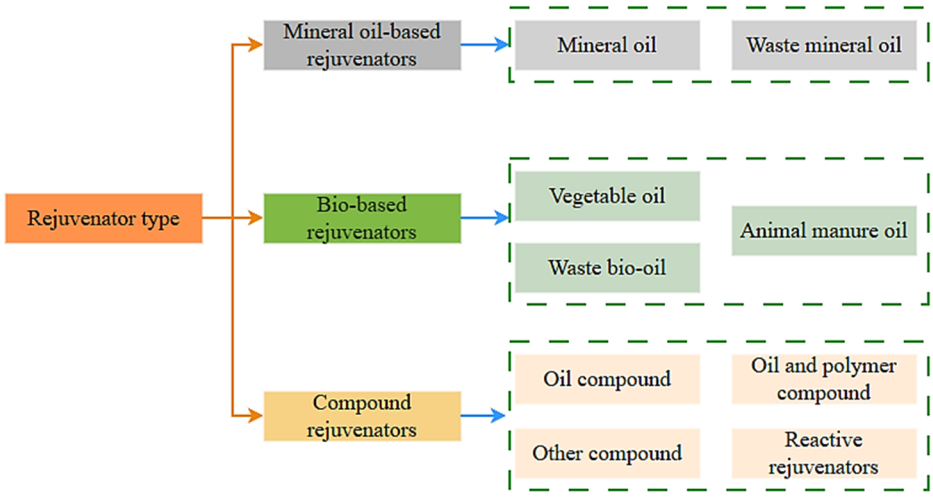


Figure 2. Classification of rejuvenators.

2.1. Mineral Oil-Based Rejuvenators

Mineral oil-based rejuvenators are by-products derived from the processing of mineral resources. Based on their raw material sources, there are two main categories: mineral oil and waste mineral oil rejuvenators.

2.1.1. Mineral Oil

Mineral oil rejuvenators are primarily derived from the products and by-products of crude oil, including AO, paraffin oil, furfural extraction oil, coke oven oil, naphthenic oil, and lubricating oil. Luo et al. [33] investigated furfural extraction oil, a by-product of lubricant production, which was enriched in cycloalkanes, monocyclic aromatics, and diaromatics. Similarly, Bajaj et al. [34] extracted AO and paraffinic oil from crude oil, which was primarily composed of polar aromatic compounds, such as unsaturated bonds. Asadi et al. [35] analyzed the compositional fractions of commercially available aromatic rejuvenators, revealing that they consisted mainly of aromatics, resins, and saturates, with aromatics as the dominant component. These findings suggest that the primary constituents of mineral oil rejuvenators include aromatics, alkanes, benzene ring compounds, and other light components. These light components effectively disperse agglomerated asphaltenes, enhancing the molecular mobility of rejuvenated asphalt and improving its overall performance.

2.1.2. Waste Mineral Oil

Waste mineral oil rejuvenators are primarily derived from waste generated in machining and automotive maintenance processes, such as WEO and waste engine oil bottoms. Wang et al. [36] delimited WEO as a solid–liquid coexisting substance formed when en-

gine oil undergoes aging processes, such as oxidation and metal abrasion, during use. Liu et al. [37] conducted an analysis of the chemical composition of WEO and found that it mainly consisted of low molecular weight compounds, including 1,3,5-trimethyl-benzene, 2,4-dimethylstyrene, 1,2,4-trimethyl-benzene, 1-ethyl-3-methyl-benzene, and 1-phenyl-1-butene. Additionally, Al-Saffar et al. [38] found that compared to engine oil, WEO exhibited a more complex composition, containing gasoline fractions, additives, and metallic elements such as Fe, Cd, Cr, Pb, Zn, and Mg, which could influence the rejuvenation performance. Due to these metallic residues, Tian et al. [39] discovered that WEO accelerated the aging of rejuvenated asphalt, thereby compromising its long-term performance. Similarly, Luo et al. [40] reported that certain metallic elements in WEO catalyzed oxidation and condensation reactions in asphalt, further accelerating its aging process. In contrast, Qiu et al. [41] analyzed residual impurities in WEO after refining and found that the chemical composition of waste engine oil bottoms closely resembled that of asphalt, making it a promising candidate for rejuvenation applications.

2.2. Bio-Based Rejuvenators

Bio-based rejuvenators are produced from a variety of biologically produced light oil or extracts, which are classified as environmentally friendly and sustainable alternatives for asphalt rejuvenation [42]. These rejuvenators are sourced from both organic biomass produced by plants or animals and from by-products of the oil and grease industry, such as WCO and grease waste. Based on the material types and their respective sources, bio-based rejuvenators are typically classified into three categories: vegetable oil, animal manure oil, and waste bio-oil. Vegetable oil rejuvenators are commonly produced from plant-derived feedstocks, including wood, algae, soybeans, and straw. Animal manure oil rejuvenators, on the other hand, are predominantly derived from animal waste products, such as pig manure and cow dung. Lastly, waste bio-oil rejuvenators, including WCO, GO, and other waste fats, serve as alternative feedstocks for asphalt rejuvenation.

2.2.1. Vegetable Oil

Vegetable oil rejuvenators can be extracted from plant materials through a variety of methods, such as physical processing (such as pressing and hammering) and chemical conversion (including fast pyrolysis [43], catalytic cracking [44], etc.). Physical processing involves the extraction of vegetable oil by mechanically squeezing or pressing plants such as tung tree fruits [45], mustard seeds [46], cotton seeds [47], and various other oil-bearing crops including olive [48], mahua [49], and SO [50]. A Gas Chromatography–Mass Spectrometry analysis of mustard press oil was carried out by Ahmad et al. [46], revealing that it predominantly consisted of non-polar erucic acids, which played a vital role in recovering the balance between asphaltenes and maltenes during the rejuvenation of asphalt. Yan et al. [45] reported that TO contained nine distinct fatty acids, with oleic acids and linolenic acids being the most abundant. Additionally, Pradhan et al. [49] identified that mahua oil contained palmitic, stearic, linoleic, and oleic acids, with oleic and linoleic acids being the most prevalent.

In addition, chemical processing methods are also widely employed in the extraction of vegetable oil. As illustrated in Figure 3, rapid pyrolysis was considered one of the most efficient techniques for producing vegetable oil. The primary products of fast pyrolysis included liquid (vegetable oil), solid (vegetable char), and gas phases [51]. Alvarez et al. [52] performed pyrolysis of rice husks at 450 °C and achieved a 70 wt.% vegetable oil yield. Similarly, Anisa et al. [53] extracted 47.3 wt.% vegetable oil from palm husks at 500 °C. Existing studies demonstrated that vegetable oil can also be efficiently extracted from various waste biomasses, such as waste wood [54], olive pomace [55], rice husk ash [56], and

biodiesel residue [57] through fast pyrolysis. Yang et al. [58] found that vegetable oil derived by fast pyrolysis was primarily composed of compounds such as 2,6-dimethoxyphenol, 2-methoxy-4-vinylphenol, and 2-cyclopentanedione, as well as alcohols, phenols, and ketones. In a similar study, Alvarez et al. [52] analyzed rice husk-derived bio-oil using Gas Chromatography–Mass Spectrometry and found that it predominantly consisted of oxygenated compounds, including phenols, cyclopentenones, furans, and aldehydes.

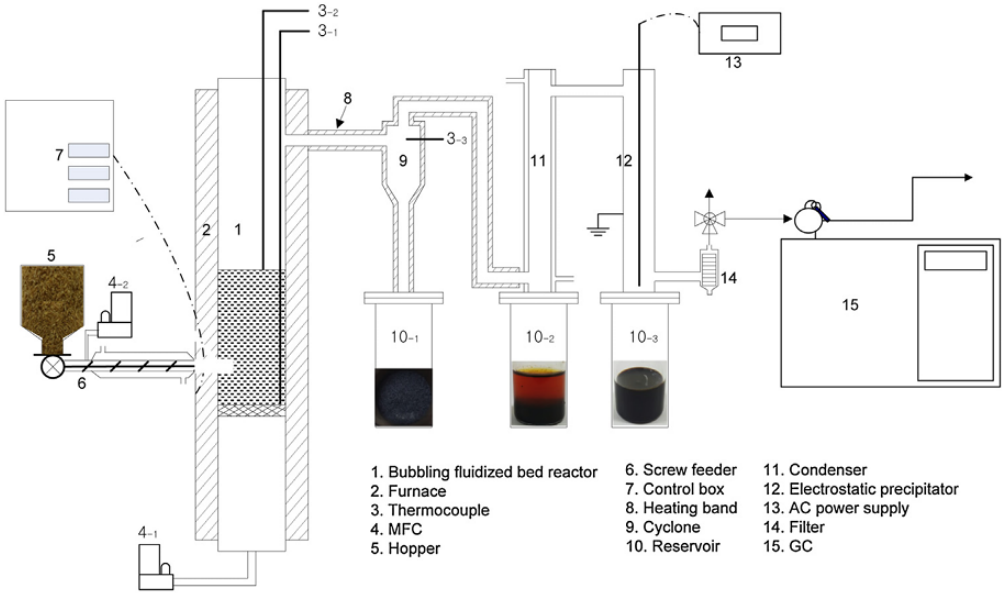


Figure 3. Schematic diagram of the bubbling fluidized bed reaction [59].

2.2.2. Animal Manure Oil

Animal manure oil rejuvenators are light oils extracted from animal manure, which is a biomass rich in water content. The oil is typically extracted through hydrothermal liquefaction and rapid pyrolysis processes [60]. Mills-Beale et al. [61] performed pyrolysis of swine manure at 380 °C to extract animal manure oil. Similarly, Fini et al. [62] subjected swine manure to pyrolysis at a temperature of 304 °C for 80 min, yielding 70 wt.% animal oil. Gas Chromatography–Mass Spectrometry analysis of oil extracted from pig manure revealed that it primarily consisted of hexadecane amide, dimethyl ketone oxime, octadecane amide, and octadecanoic acids [62]. This analysis indicated that animal manure oil is rich in nitrogen, which is primarily associated with the amide groups present. Additionally, amide groups were found by Fini et al. [62] to be capable of efficiently depolymerizing agglomerated asphaltenes, which would restore the molecular structure of aged asphalt. Oldham et al. [63] reported that oil derived from the manure of pigs could increase the stiffness and viscosity of aged asphalt, thereby improving overall performance.

2.2.3. Waste Bio-Oil

Similarly to waste mineral oil, waste bio-oil also represents a significant stockpile of rejuvenators. Waste bio-oil rejuvenators are primarily derived from waste vegetable oil and animal manure oil, which are generated from restaurants, cafeterias, and food processing facilities. Waste vegetable oil predominantly includes WCO, GO, and similar by-products, which consist of fresh oil mixed with various impurities after use. The physical properties of waste vegetable oil can change during usage, and some waste edible vegetable oil rejuvenators may undergo a liquid-to-solid phase transformation at a relatively low temperature, thereby affecting the rejuvenation efficiency [14]. Several researchers analyzed the chemical components of waste sunflower oil and identified that waste sunflower oil primarily contains linoleic acid (55.54%), oleic acid (30.56%), and palmitic acid (9.94%) [64].

The major component of WCO is free fatty acids, which include oleic acids, linoleic acids, palmitic acids, and stearic acids [19]. Based on the research conducted by Ahmed et al. [9], oleic acids and palmitic acids—two saturated fatty acids—are hydrophilic in nature. This hydrophilicity contributed to the high moisture sensitivity of rejuvenated pavements treated with vegetable oil, which accelerated early-stage damage to asphalt pavements.

Waste animal oil predominantly includes waste fish oil, waste lard, and other animal-derived oil generated during processing. Wiggers et al. [65] employed rapid pyrolysis to treat waste fish oil at 525 °C and subsequently extracted light and heavy oil through distillation. Sang et al. [66] reported that waste lard could significantly enhance both the toughness and adhesion of aged asphalts, thereby improving the overall performance.

2.3. Compound Rejuvenators

In recent research on rejuvenators, the combined influence of various categories of rejuvenators has been the subject of considerable attention. Studies demonstrated that bio-based and mineral oil-based rejuvenators effectively restored the low-temperature rheological properties of aged asphalt [67–70]. However, their influence on improving the high-temperature deformation resistance and repairing degraded polymers in aged SBSMA remained limited. To address these challenges, several researchers incorporated polymers, resulting in the development of new compound rejuvenators [71–74]. Moreover, Xu et al. [75] introduced the concept of reactive rejuvenators specifically designed for aged SBSMA. This rejuvenator simultaneously repaired both degraded SBS modifiers and aged asphalt, leading to substantial improvements in the properties of SBSMA. Additionally, Zhang et al. [76] examined the composite use of a viscosity enhancer and nitric acid solution, which also exhibited promising rejuvenation effects. Based on functional characteristics and composition, compound rejuvenators can be classified into four categories: oil compound rejuvenators, oil and polymer compound rejuvenators, reactive rejuvenators, and other compound rejuvenators.

2.3.1. Oil Compound Rejuvenators

There are various types of compound rejuvenators, including multi-component combinations of bio-based rejuvenators, mineral oil-based rejuvenators, or a combination of bio-based and mineral oil-based rejuvenators. Commonly used composites include combinations such as cashew nut phenol with distilled tall oil, algal extracts with lipid-rich animal manure oil, and CO with coke oven oil. Ye et al. [77] examined the rejuvenation effect of a combined application of cashew nut phenol oil and distilled tallow oil, finding that this combination effectively softened asphalt and depolymerized asphaltenes. Similarly, Pahlavan et al. [78] discovered that the combination of algae and swine manure was effective in breaking down asphaltenes into smaller fractions, which were subsequently dispersed in maltene. This process not only reduced asphalt agglomeration but also enhanced water stability. Overall, oil-based compound rejuvenators exhibit similar effects to single oil-based rejuvenators.

2.3.2. Oil and Polymer Compound Rejuvenators

The compounding of oil-based rejuvenators with polymers has been applied and has attracted considerable research interest due to its enhanced rejuvenation effectiveness. Commonly used polymers include scrap rubber [71], SBS [23,79,80], and Styrene-Butadiene Rubber (SBR) [81]. Eltwati et al. [80] discovered that AO softened aged asphalt and improved rejuvenated asphalt's low-temperature properties, but the high-temperature properties of the rejuvenated asphalt were compromised, while SBS modifiers managed to improve the properties of the rejuvenated asphalt at high temperatures. Furthermore, Yao et al. [82] found, through molecular dynamics simulations, that SBS modifiers exhibited enhanced

compatibility with asphalt when the aromatic content was high and the asphaltene content was low. This indicated that SBS modifiers combined with light oil may better enhance the properties of asphalt that has been rejuvenated. Li et al. [83] observed that SBR exhibited outstanding performance in improving fatigue cracking resistance and low-temperature cracking resistance of RAP. To determine the optimal SBS addition method, Dong et al. [84] compared wet and dry SBS incorporation methods in rejuvenated asphalt mixtures. They found that dry SBS effectively diffused into aged asphalt, replenishing the fractured and aged SBS. The polymer counteracted the adverse effects of light oil on the high-temperature properties of aged asphalt, thereby enhancing the overall rejuvenation effect. Notably, degraded polymer in aged modified asphalt remains unrepaired or unused, leading to resource wastage.

2.3.3. Reactive Rejuvenators

SBSMA is widely utilized due to its exceptional performance. However, the rejuvenation of SBS after aging remains a significant technical challenge. Traditional rejuvenation methods are insufficient for effectively repairing degraded SBS in aged SBSMA, as they primarily replenish light components. Xu et al. [24] identified that fractured SBS chains consisted mainly of hydroxyl and carboxyl chain segments. These segments could re-establish a cross-linked structure through a ring-opening reaction with epoxy compounds or an addition reaction with isocyanate compounds. The repair process is illustrated in Figure 4. Currently, reactive rejuvenators are composed of reactive compounds and light oil fractions. Based on the structural characteristics of the reactive compounds, reactive rejuvenators can be classified into two categories: the epoxy system and isocyanate system.

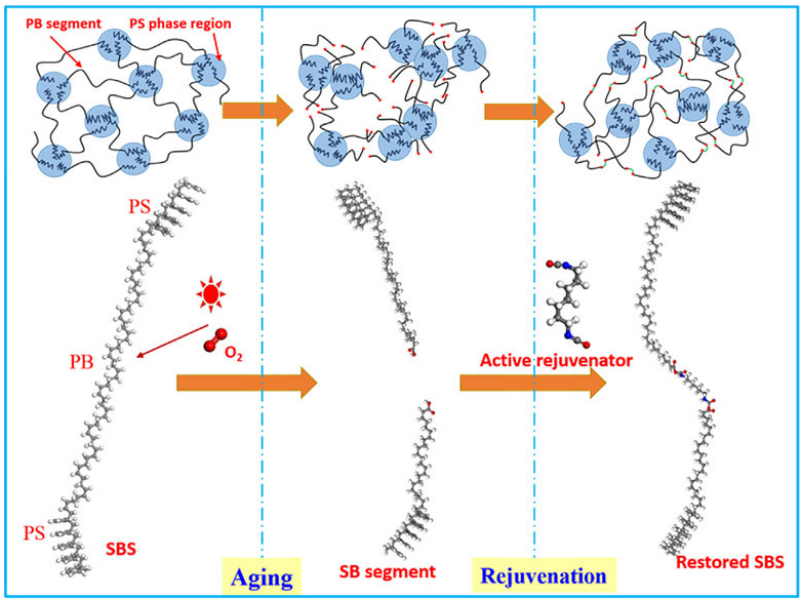


Figure 4. Diagram for structural destruction and reconstruction of modifier SBS [85].

(a) Epoxy system

The epoxy system refers to a compound rejuvenator composed of epoxy compounds, catalysts, and light oil. In this system, epoxy compounds undergo a ring-opening reaction with the broken SBS molecular chain ends, facilitating chemical crosslinking that restores the original network structure of aged SBSMA. This reaction is typically catalyzed. Concurrently, light oil enhances the solubility and fluidity of epoxy compounds while also regulating the components of aged virgin asphalt [24]. It can be inferred from the structural characteristics of the compounds that compounds with benzene rings or short-chain structures are stable at high temperatures, thereby increasing asphalt stiffness, whereas

compounds containing long-chain structures contribute to improving asphalt flexibility [86]. Common epoxy compounds include ESO [87], BUDGE [88], TMPGE [89], HDDGE [88], and TGDOM [89], among others. Xu et al. [88] found that BO softened aged asphalt, while HDDGE promoted the reaction between the carboxyl group in the fractured SBS chain and its epoxy group through a ring-opening reaction. This ring-opening reaction served as a key rejuvenation mechanism for repairing degraded SBS molecules. Wei et al. [90] reported that using TPP as a catalyst could open the epoxy groups in epoxy compounds, facilitating their interaction with characteristic groups of fractured SBS. FTIR tests revealed that the increase in ether and ester peaks indicated that the epoxy groups reacted with fractured SBS, effectively reconstructing the SBS lattice structure. Furthermore, Xu et al. [91] concluded that the tri-epoxy structure of TMPGE was more effective than the bi-epoxy structure of BUDGE in restoring the macroscopic properties of aged SBSMA. Chiang et al. [92] further explored the bi-epoxy structure of di-epoxy HDE, which was found to repair degraded SBS. Relatively, the mono-epoxy structure of the rejuvenator only had a physical softening effect and did not chemically repair SBS.

(b) Isocyanate system

The isocyanate system is a kind of compound rejuvenator consisting of isocyanate compounds and light oil. In this system, highly reactive isocyanate groups can quickly chemically react with the broken SBS molecular chain. Common compounds used in isocyanate system include MDI [93], 1,6-Diisocyanatohexane (HDI) [94], and TDI [94], among others. Research conducted by Xu et al. [75] demonstrated that combining isocyanate compounds with light oil improved the dispersion and fluidity of aged SBSMA. A more comprehensive recovery of asphalt properties was achieved by enhancing the penetration ability of the reactive compounds into the molecular chains of aged asphalt. Additionally, Zhang et al. [95] found through quantum chemical simulations that the reactions between TDI and HDI with certain reactive groups of aged SBSMA occurred spontaneously, without the need for a catalyst, which demonstrated the high reactivity of the isocyanate system. However, different isocyanate compounds exhibited varying reaction efficiencies with hydroxyl or carboxyl groups and thus should be selected based on specific requirements. Unlike the epoxy system, the isocyanate system reacts with the broken SBS chain via nucleophilic addition reactions at the hydroxyl segment and polycondensation (amine esterification) at the carboxyl segment, resulting in a more efficient restoration of the properties of aged SBSMA [96,97].

Interestingly, the epoxy-reactive rejuvenator primarily focuses on enhancing the low-temperature flexibility of aged SBSMA, while the isocyanate-reactive rejuvenator aims to improve the high-temperature properties [24]. Therefore, Xu et al. [98] combined epoxy compounds with isocyanate compounds, which not only suppressed the decline in the softening point of aged SBSMA but also improved the low-temperature toughness of the rejuvenated asphalt.

2.3.4. Other Compound Rejuvenators

In addition to the commonly used compound rejuvenators, there are several specialized rejuvenators with distinct roles. For instance, a compound rejuvenator consisting of light oil, a viscosity builder, and dilute nitric acid could provide better recovery for the adhesive properties of aged asphalt [76]. WCO pretreated with a plasticizer could mitigate the excessive softening influence of WCO. So, it could improve the high-temperature rutting resistance of rejuvenated asphalt [99]. A compound rejuvenator made from mineral oil, epoxy resin, and a curing agent could enhance the properties of aged asphalt in terms of low-temperature properties, toughness, and viscosity [100]. Furthermore, Hu et al. [101] discovered that PU could improve the compatibility of rejuvenators with asphalt. The com-

pound rejuvenator, consisting of PU and BUDGE, enhanced the low-temperature properties and flexibility of aged SBSMA.

3. Micro-Properties of Rejuvenated Asphalt

3.1. Surface Micro-Structure

Currently, numerous academics have extensively examined the microstructure of rejuvenated asphalt. Common research instruments include AFM, FM, and SEM, which are applied to analyze, from a microscopic standpoint, the mechanism of action between rejuvenators and aged asphalt.

3.1.1. Atomic Force Microscope

At present, researchers usually apply AFM to study the microstructure of asphalt. Chen et al. [102] found that whether the rejuvenator reacted with aged asphalt could be characterized by changes in the microstructure of the asphalt surface. As shown in Figure 5, there was no significant change in the number and size of "bee structures" in rejuvenated asphalt after the addition of a high-aromatic rejuvenator. This suggested that the microstructure of rejuvenated asphalt was not affected by the high-aromatic rejuvenator [102]. Rafiq et al. [103] discovered that new "dark structures" were formed in the creation of rejuvenated asphalt with the addition of crude palm oil, as shown in Figure 6. Building on this, Jiang et al. [87] found that a compound rejuvenator consisting of CO and ESO could recover the quantity of "bee structures" and reduce their average length in rejuvenated asphalt. Furthermore, a compound rejuvenator composed of aromatic oil, viscosity enhancers, and concentrated nitric acid produced "gully structures" and "hole structures" which were new on the surface of rejuvenated asphalt. These structures made rejuvenated asphalt rough by reducing the smoothness of its surface [76]. A compound rejuvenator comprising a warm mix agent and light oil made the "bee structures" on rejuvenated asphalt's surface more evenly distributed, thereby reducing its roughness [104]. Overall, the addition of rejuvenators induced varying degrees of changes in the micro-properties of rejuvenated asphalt. Unfortunately, research regarding the micro-properties of compound-based rejuvenated asphalt observed through AFM remains in a state of deficiency. The impact of mixing multiple compounds on the "bee structures" of aged SBSMA remains an area for further investigation.

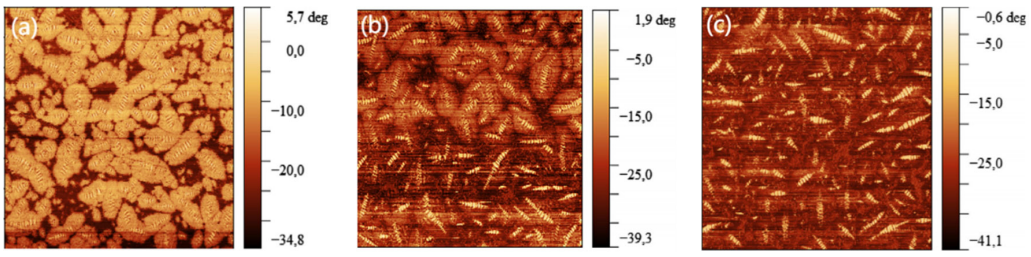


Figure 5. AFM diagram of different asphalt: (**a**) virgin asphalt, (**b**) aged asphalt, (**c**) rejuvenated asphalt [102].

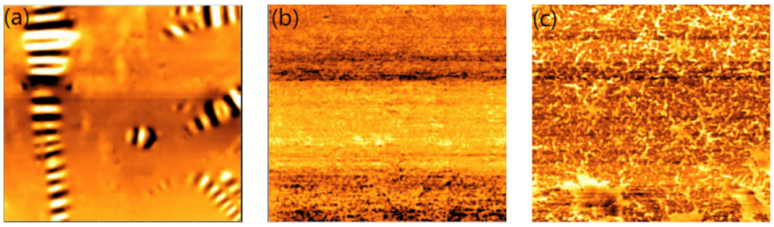


Figure 6. AFM diagram of different asphalt: (**a**) virgin asphalt, (**b**) aged asphalt from RAP, (**c**) rejuvenated asphalt [103].

3.1.2. Fluorescence Microscope

FM is primarily used to observe changes in the internal structure of aged and rejuvenated asphalt. Ding et al. [105] concluded that aging led to a decrease in fluorescence intensity of asphalt, which could be used to differentiate aged asphalt from virgin asphalt by observing changes in fluorescence intensity. When conducting research on the aging and rejuvenation processes of modified asphalt, noticeable spot changes can indicate the effect of the rejuvenator on aged modified asphalt. It can be observed from Figure 7 that the number of spots in the asphalt FM diagram increase significantly with the increase in BO doping, and the spots are uniformly dispersed [106]. This was attributed to the molten aromatic structure of BO, which promoted the homogeneous dispersion of BO in the rejuvenated asphalt [107]. When a compound rejuvenator consisting of mineral oil, epoxy resin, and curing agent was added, mineral oil could reduce the viscosity of rejuvenated asphalt and provide more reaction space for epoxy resin's curing reaction [100]. Additionally, as shown in Figure 8, Xu et al. [88] carefully analyzed the difference in the FM diagrams of BO- and epoxy-system-rejuvenated asphalt and found that the epoxy system rejuvenator was superior to the bio-based rejuvenator in terms of spot recovery. Cao et al. [85] compared the effect of a combination of BO and isocyanate compounds (HDI, MDI, and TDI) on aged SBSMA and discovered that isocyanate compounds rebuilt some of the damaged SBS linkages. The FM diagram of the rejuvenated asphalt showed noticeable cross-linking phenomena. Xu et al. [24] further compared the FM diagram of epoxy system and isocyanate system rejuvenators and found that both recovered the broken segments and homogeneity of aged SBSMA more effectively.

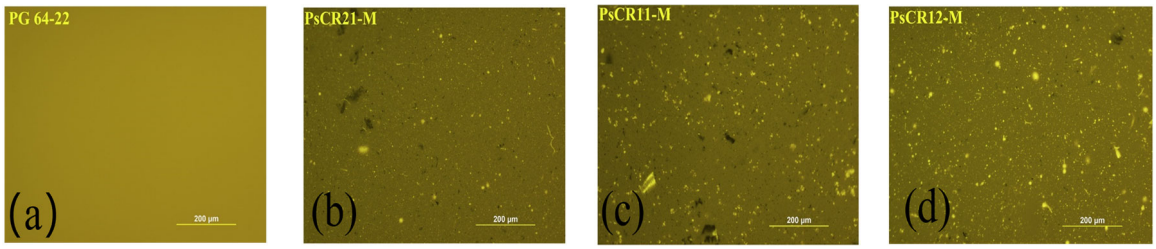


Figure 7. FM diagram of different samples: (**a**) virgin asphalt, (**b**) asphalt with 10% BO, (**c**) asphalt with 15% BO, (**d**) asphalt with 20% BO [106].

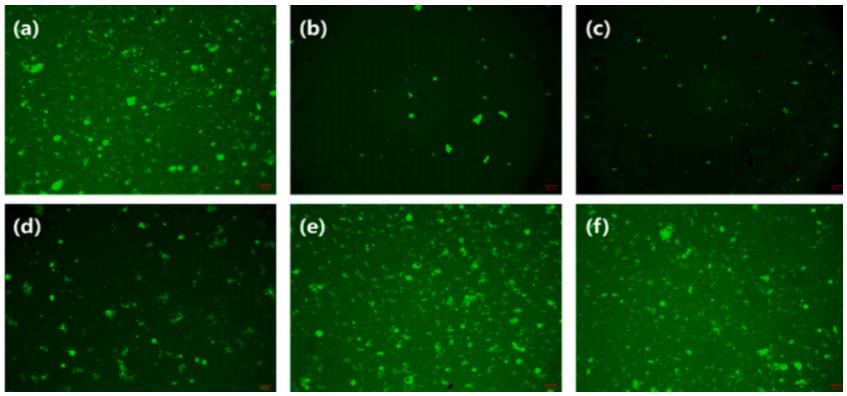


Figure 8. FM diagram of different samples: (**a**) SBSMA, (**b**) aged SBSMA, (**c**) BO-rejuvenated SBSMA, (**d**) BO-, HDDGE-, RDGE-, and BDMA-rejuvenated SBSMA, (**e**) BO-, HDDGE-, and PEHA-rejuvenated SBSMA, (**f**) BO-, HDDGE-, RDGE-, and PEHA-rejuvenated SBSMA [88].

3.1.3. Scanning Electron Microscope

When it comes to SEM, it is usually applied to analyze the surface morphology of asphalt and its compatibility with rejuvenators by comparing the roughness, flatness, and

texture state of the surface topography [108]. It can be seen from Figure 9 that the surface morphology of rejuvenated asphalt can be improved by adding CO. In addition, when GO was added, the surface of the rejuvenated asphalt exhibited large folds and an uneven condition, indicating a poor repair effect [109]. In contrast, BO was able to restore the cracked areas, forming a continuous structure [110]. Furthermore, Li et al. [111] found that compound rejuvenators consisting of WEO and WCO successfully integrated into the grooves on the asphalt surface. Further, Xu et al. [100] found that compound rejuvenators consisting of a curing agent, epoxy compounds, and mineral oil resulted in a porous three-dimensional mesh structure, weakening the association compactness between epoxy compounds and asphalt.

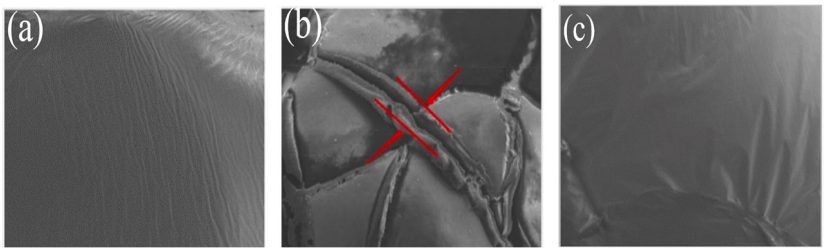


Figure 9. SEM diagrams (20.0 μm) of different samples: (**a**) virgin asphalt, (**b**) aged asphalt, (**c**) CO-rejuvenated asphalt [109].

3.2. Functional Groups

FTIR is widely used in the research of functional groups in asphalt, providing a clear understanding of the chemical reactions between aged asphalt and rejuvenators [112]. Among these, the representative aging functional groups in asphalt are the carbonyl group (C=O), sulfoxide group (S=O), and olefin group (C=C). Additionally, to quantitatively characterize the changes in functional groups when rejuvenating aged asphalt, the carbonyl index ($I_{C=O}$) and sulfoxide index ($I_{S=O}$) were utilized to assess the aging degree of asphalt [113]. The formulas for calculating $I_{C=O}$ and $I_{S=O}$ are presented in Formula (1) and Formula (2), respectively. In the case of aged SBSMA, the repair of degraded SBS may generate C=O or S=O, making traditional $I_{C=O}$ and $I_{S=O}$ insufficient for accurately analyzing changes in SBSMA. Therefore, the structural damage index (BSI) is commonly used to evaluate SBS degradation, with its formula provided in Formula (3) [114].

$$I_{S=O} = \frac{Characteristic\ peak\ area\ of\ sulfoxide\ group\ near\ 1030\ cm^{-1}}{Peak\ zone\ area\ between\ 2000\ cm^{-1}\ and\ 600\ cm^{-1}} \quad (1)$$

$$I_{C=O} = \frac{Aromatic\ characteristic\ peak\ area\ near\ 1600\ cm^{-1}}{Peak\ zone\ area\ between\ 2000\ cm^{-1}\ and\ 600\ cm^{-1}} \quad (2)$$

$$BSI = \frac{characteristic\ peak\ area\ of\ polybutadiene\ segment\ near\ 966\ cm^{-1}}{characteristic\ peak\ area\ of\ polystyrene\ segment\ near\ 699\ cm^{-1}} \quad (3)$$

By analyzing the $I_{C=O}$ and $I_{S=O}$ of different rejuvenated asphalts in recent years, the chemical changes in rejuvenated asphalt can be analyzed. As shown in Figure 10, the increase in $I_{C=O}$ was noticeable when asphalt aged. This phenomenon implies that the aging products of asphalt contain higher levels of carbonyl components. When a mineral oil-based rejuvenator was added, the $I_{S=O}$ of the rejuvenated asphalt decreased, though at varying rates. For instance, AO reduced both $I_{C=O}$ and $I_{S=O}$ by dissolving some oxidation products, such as asphaltenes. When maltene was added, the $I_{C=O}$ and $I_{S=O}$ were slightly reduced due to maltene's lower content of oxygen-containing functional groups, which slowed the oxidation process in rejuvenated asphalt [115]. The addition of WEO increased $I_{S=O}$ and decreased $I_{C=O}$ in rejuvenated asphalt, with the effect becoming more pronounced

as the rejuvenated asphalt aged. This was because the scrap metal in WEO promoted the oxidation of rejuvenated asphalt, leading to a continuous increase in $I_{S=O}$, while the light components in WEO reduced $I_{C=O}$ [40]. Therefore, mineral oil can well reduce $I_{C=O}$ and $I_{S=O}$ and recover the properties of aged asphalt, while waste mineral oil may influence the road application of rejuvenated asphalt.

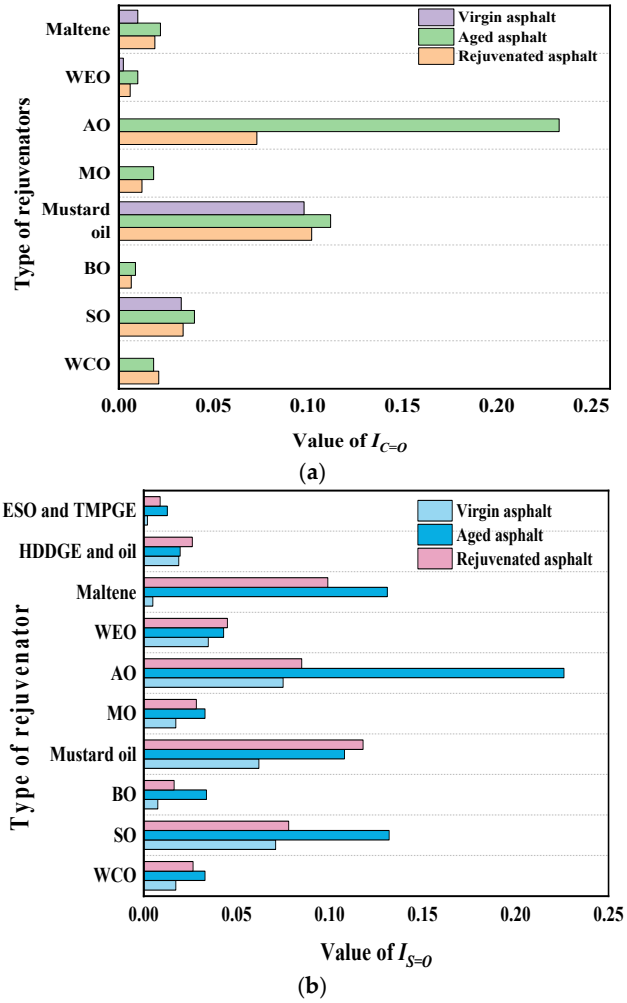


Figure 10. The results of FTIR: (**a**) the value of $I_{C=O}$, (**b**) the value of $I_{S=O}$ [70,115–122].

When a bio-based rejuvenator was added, SO caused both the $I_{C=O}$ and $I_{S=O}$ of the rejuvenated asphalt to decrease, with a significant reduction observed. However, when WCO was added, $I_{S=O}$ showed only a slight decrease, while $I_{C=O}$ increased instead of decreasing. Cao et al. [69] and Zhang et al. [121] attributed the increase in $I_{C=O}$ to the reaction of oleic, fatty, and palmitic acids present in WCO with oxygen. Further, Zahoor et al. [5] found that with a little WCO added to aged asphalt, $I_{S=O}$ and $I_{C=O}$ were effectively decreased. Although the light components in WCO could balance the colloidal components in rejuvenated asphalt, its rejuvenating effect was not as pronounced as that of fresh vegetable oil.

In addition, when a reactive rejuvenator consisting of ESO and TMPGE was added to aged SBSMA, a decrease in $I_{S=O}$ was observed. This indicated that the ESO restored the aged asphalt, but it did not restore the aged SBSMA [122]. To better analyze the rejuvenation of aged SBSMA, Zhang et al. [93] investigated the change in polybutadiene content in MDI-rejuvenated SBSMA. The experiment made clear that the BSI rose with the increasing rate of MDI dosage, indicating that MDI effectively repaired the broken SBS chains and re-established the crosslinked mesh structure.

3.3. Molecular Weight

GPC is currently the primary research tool to analyze the changes in molecular weight in asphalt. It allows for the determination of data such as number-average molecular weight (M_n) and weight-average molecular weight (M_w) [123]. They characterized changes in small and large molecules [123]. It can be obtained from Figure 11 that both M_n and M_w values increased as the asphalt aged. When rejuvenators supplemented with light components, such as mineral oil-based and bio-based rejuvenators, were added, M_w in the rejuvenated asphalt significantly decreased. This suggested that rejuvenators containing light components could restore the number of large molecules in rejuvenated asphalt through a "dilution" effect. In contrast, degradation of SBS occurred when SBSMA aged [124]. Therefore, it is crucial to consider not only molecular weight changes in virgin asphalt but also changes in the molecular weight of the degraded SBS modifier during the analysis. When BO was added, the number of large molecules in the rejuvenated SBSMA increased, which was inconsistent with the previous conclusion. Jiang et al. [87] investigated this phenomenon and found that there was a high content of unsaturated fatty acid groups in BO, which caused a sharp increase in molecular weight at the initial stage of rejuvenator addition. However, after the rejuvenated asphalt was exposed to normal temperatures for 7 days, it was found that the molecular weight was lower than that of the virgin asphalt. Because the unsaturated fatty acids reacted with polar large molecules in the rejuvenated asphalt, it contributed to a change in molecular weight. Additionally, in the asphalt rejuvenated by reactive compounds, epoxy compounds recombined with some of the broken SBS chains, which was reflected by a leftward shift in the polymer peaks. This could recover molecular weight of rejuvenated SBSMA in comparison to virgin sample, which made it clear that the broken SBS chains in the rejuvenated SBSMA had been repaired [122].

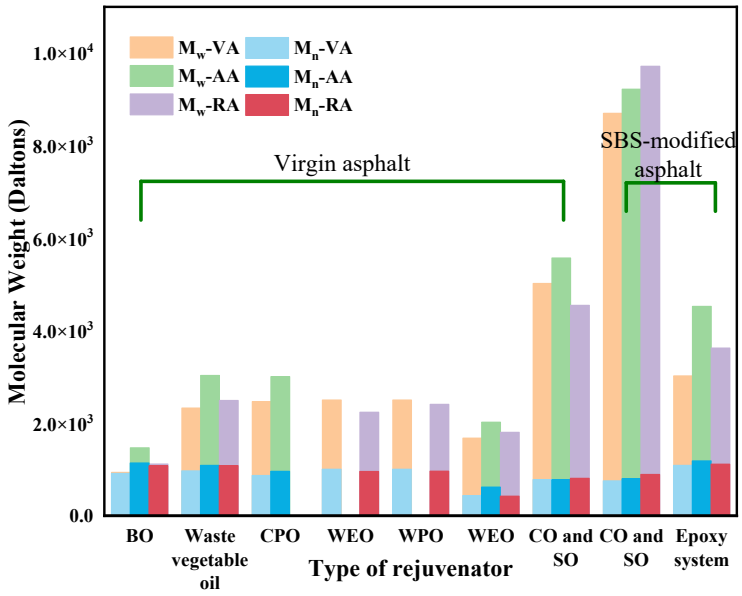


Figure 11. The molecular weight of virgin asphalt, aged asphalt, and rejuvenated asphalt [69,70,87,122,125,126].

4. Macro-Properties of Rejuvenated Asphalt

4.1. Three Indicators

Penetration, softening point, and ductility are ordinarily applied to assess the basic properties of asphalt. This review summarizes recent research on the evaluation of recovery effect of different rejuvenators on three indicators of asphalt.

4.1.1. Penetration

The penetration of asphalt primarily reflects changes in its hardness. Figure 12 illustrated the recovery action of bio-based, mineral oil-based, and compound rejuvenators on the penetration of rejuvenated asphalt. As asphalt ages, its penetration decreases due to the increase in asphaltenes [109]. It is evident that different types of rejuvenators restore the penetration of rejuvenated asphalt close to that of virgin samples. With sufficient dosage, rejuvenators can even improve the penetration of rejuvenated asphalt. This shows a softening effect on the rejuvenated asphalt. Regarding the rejuvenation of aged SBSMA, when adding oil and polymer compound rejuvenators as well as reactive rejuvenators to it, they can supplement or repair the degraded SBS and enhance the asphalt's performance. This rejuvenation effect is superior to that achieved by adding only light oil rejuvenators [90].

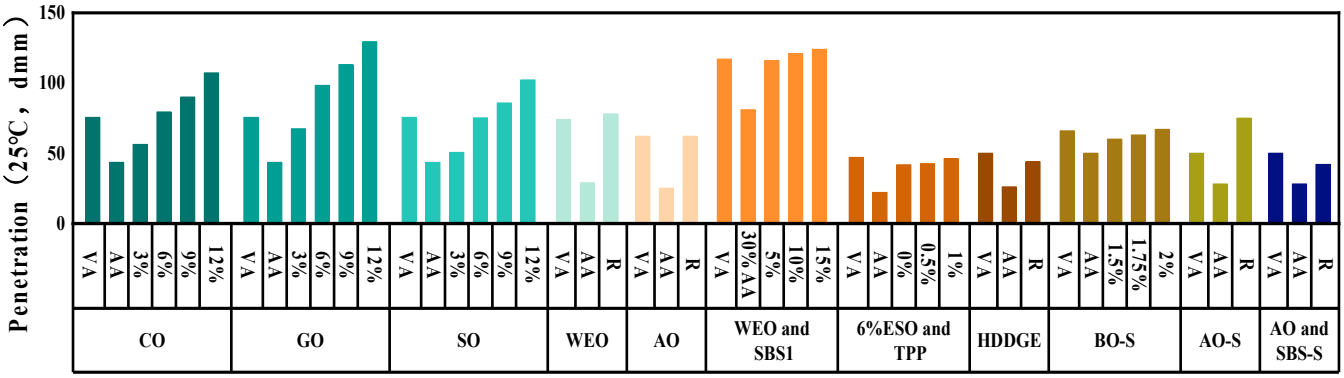


Figure 12. Penetration of virgin asphalt (VA), aged asphalt (AA), and rejuvenated asphalt (RA) [23,57,70,79,90,109,116,127].

4.1.2. Softening Point

The softening point reflects changes in the asphalt's high-temperature stability. It can be seen from Figure 13 how bio-based, mineral oil-based, and compound rejuvenators, respectively, affect the softening point of rejuvenated asphalt. Obviously, the softening point increases with asphalt aging. However, Hong et al. [23] found that the softening point decreased with SBSMA aging because the degradation of the SBS polymer and the destruction of the crosslinked structure contributed to a reduction in high-temperature properties of SBSMA. The softening point of rejuvenated asphalt decreased when adding rejuvenators, effectively softening the asphalt. However, when WCO and MDI compounds were added to aged SBSMA, the softening point gradually increased, which can be attributed to the structure of the rejuvenators [128].

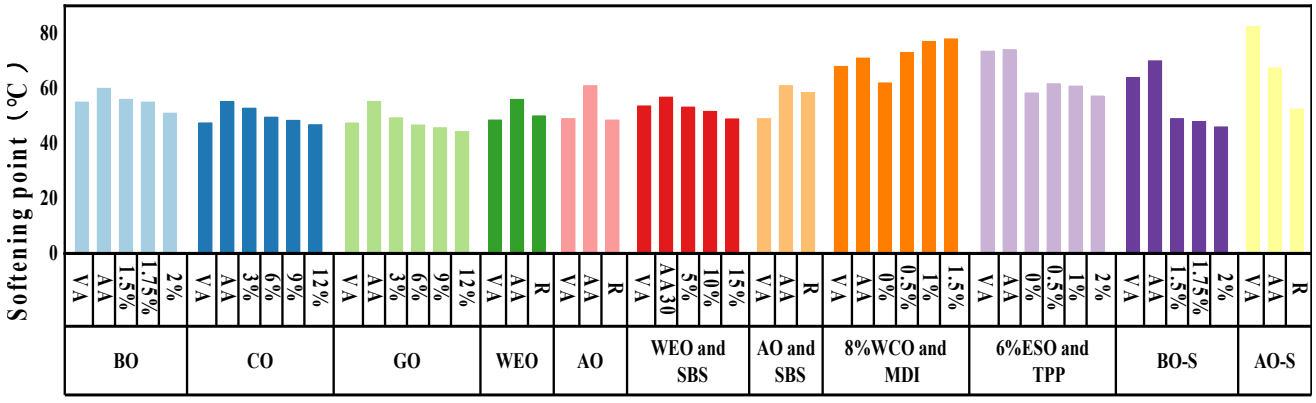


Figure 13. Softening point of virgin asphalt (VA), aged asphalt (AA), and rejuvenated asphalt (RA) [23,57,70,79,90,93,109,116,127].

4.1.3. Ductility

Ductility mainly reflects changes in flexibility and low-temperature cracking resistance of rejuvenated asphalt. Figure 14 shows how the bio-based, mineral oil-based, and compound rejuvenators influence rejuvenated asphalt's ductility. Asphalt becomes hard and brittle through aging. This may cause the ductility to decrease dramatically [129]. The addition of rejuvenators could increase the ductility of rejuvenated asphalt. However, with an increasing dosage of WEO and WPE mixtures, the ductility of rejuvenated asphalt decreases. This is due to the fact that the addition of WPE increases the proportion of discontinuous phases in rejuvenated asphalt. This makes rejuvenated asphalt easier to crack [126]. This phenomenon indicates that the dosage of rejuvenators is not necessarily proportional to the performance of rejuvenated asphalt. Furthermore, some light oil rejuvenators and reactive rejuvenators were selected in this review to compare their rejuvenation effects on aged SBSMA. The ductility of rejuvenated SBSMA gradually increased with the increase in BO dosage [23], while MDI did not effectively recover the ductility of the rejuvenated asphalt. Moreover, when HDDGE was added, the ductility recovery effect of rejuvenated SBSMA was improved compared with that of MDI. This indicated that different reactive rejuvenators vary in their recovery effects on rejuvenated SBSMA at low temperatures. Owing to the difference in composition, the epoxy system focused more on the recovery of low-temperature properties of rejuvenated SBSMA compared to the isocyanate system [24].

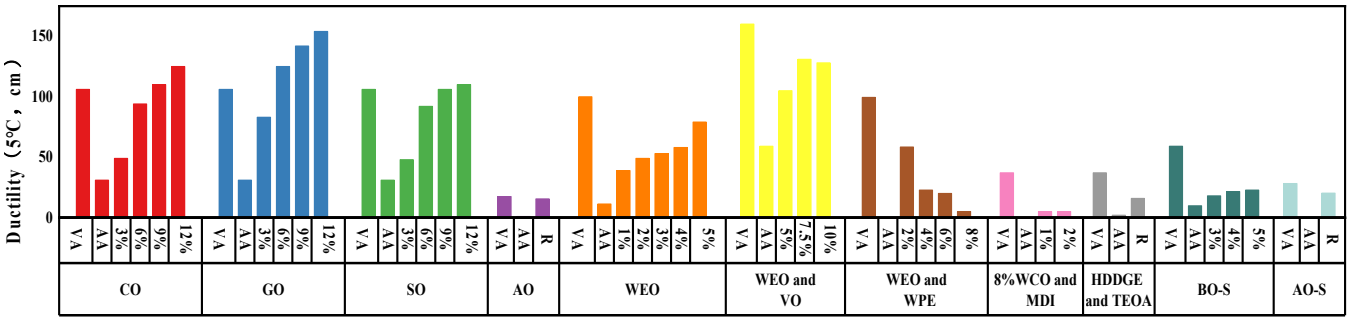


Figure 14. Ductility of virgin asphalt (VA), aged asphalt (AA), and rejuvenated asphalt (RA), waste polyethylene (WPE) [23,93,109,116,126,127,130,131].

4.2. High-Temperature Rheological Properties

Rheological properties at high temperatures are important indexes to evaluate the properties of asphalt. Chen et al. [70] discovered that the low-polar aromatic fractions of WEO were key factors in softening rejuvenated asphalt. Additionally, WEO-rejuvenated asphalt demonstrated good storage stability [70]. Similarly, Gong et al. [57] and Chen et al. [132] investigated that bio-based rejuvenators could simultaneously reduce aged virgin asphalt and aged modified asphalt's rutting factors, as well as damage the high-temperature rutting resistance of rejuvenated asphalt and SBSMA. Furthermore, Hong et al. [23] made a comparison between the effects of polymer and AO and plain AO on rejuvenated asphalt. They found that 15% polymer and AO could recover the complex shear modulus and phase angle of rejuvenated asphalt close to those of virgin asphalt, which was superior to the addition of plain AO. Eltwati et al. [79] found that a 5% compound of WEO and SBS rejuvenator significantly increased the rutting factor of rejuvenated asphalt, thus enhancing its rutting resistance. This improvement was attributed to SBS, which mitigated the excessive softening effect caused by WEO. Moreover, MDI and WCO could increase the complex shear modulus and rutting factor of rejuvenated SBSMA, thereby enhancing the high-temperature rutting resistance of rejuvenated SBSMA [93]. Further, Xu et al. [75] made a comparison between high-temperature resistance of rejuvenated asphalt with the

epoxy and isocyanate systems using the Multiple Stress Creep Recovery test and found that the non-recoverable compliance (J_{nr}) values of rejuvenated asphalt with the isocyanate system were lower than the rejuvenation effect of the epoxy system. This made it clear that the rutting resistance of isocyanate-system-rejuvenated asphalt was superior to that of epoxy-system-rejuvenated asphalt. Xie et al. [133] analyzed the rutting factor of isocyanate- and epoxy-system-rejuvenated asphalt and reached the same conclusion.

4.3. Medium-Temperature Rheological Properties

The fatigue resistance of asphalt is usually characterized by medium-temperature rheological properties [134]. Through DSR experiments, it was found that 15% AO recovered the fatigue coefficient of rejuvenated asphalt compared to that of a virgin sample across a wide temperature range [23]. Similarly, 10% waste vegetable oil was able to recover the fatigue resistance of rejuvenated asphalt to that of a virgin sample, but excessive dosing of rejuvenators can inhibit the improvement of fatigue performance in rejuvenated asphalt [69]. Furthermore, Rai et al. [135] compared the rejuvenation conditions of WCO, WEO, and TO on the fatigue resistance of rejuvenated asphalt using Linear Amplitude Sweep experiments. The researchers discovered that WEO exhibited the longest fatigue life, which indicated that it was more effective at recovering rejuvenated asphalt's fatigue resistance.

In addition, Eltwati et al. [79] tested the effects of a 5% rejuvenator composed of SBS and WEO on aged asphalt and found that a 5% compound rejuvenator significantly improved the medium-temperature fatigue resistance of rejuvenated asphalt. Similarly, a 15% blend of AO and SBS demonstrated good fatigue resistance in rejuvenated asphalt between 10 °C and 30 °C [23]. Further, Linear Amplitude Sweep-based investigations by Xu et al. [75] documented the efficacy of a AO and TAIC co-formulation in rehabilitating fatigue life (N_f) of aged SBSMA, attaining fatigue durability parity with the virgin sample. Likewise, a 1% blend of MDI and WCO achieved a balance between fatigue resistance and high-temperature resistance in rejuvenated SBSMA, showing effective rejuvenation [93]. Wei et al. [90] found that the addition of TPP enhanced the repair of the SBS crosslinked structure by ESO, resulting in an increase followed by a decrease in the complex shear modulus, which in turn improved fatigue resistance. As shown in Figure 15, compound rejuvenators consisting of coffee residue and WCO significantly improved rejuvenated asphalt's fatigue resistance performance [136]. Generally, proper amounts of different rejuvenators can improve the durability of rejuvenated asphalt against fatigue.

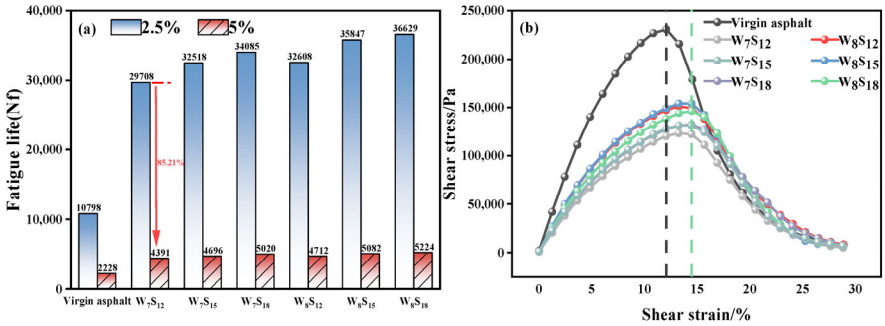


Figure 15. Fatigue life, stress–strain relationship at two strains: (**a**) fatigue life of recycled asphalt; (**b**) stress–strain relationship [136].

4.4. Low-Temperature Rheological Properties

Low-temperature rheological properties are applied to assess the cracking resistance of asphalt in a low-temperature environment. The creep rate (m) of WEO-rejuvenated asphalt was determined to be higher than the value of aged asphalt, essentially matching the creep rate of a virgin sample, as assessed through the Bending Beam Rheometer

test [70]. Further studies on performance changes in WEO-rejuvenated asphalt after storage for 10 and 30 days revealed that the temperature at which rejuvenated asphalt became damaged increased, though it remained superior to that of the virgin sample [137]. Based on this, Luo et al. [40] assessed the low-temperature cracking resistance of WEO-rejuvenated asphalt after secondary aging through direct tensile tests. They found that after 40 h of aging, the low-temperature cracking resistance of the aged asphalt was severely degraded, which was not conducive to practical applications. Additionally, Hong et al. [23] compared the recovery of low-temperature properties in rejuvenated virgin asphalt and SBSMA when adding AO, discovering that the recovery in rejuvenated SBSMA was better than that of rejuvenated virgin asphalt. Similarly, Lv et al. [138] added different dosages of vegetable oil to aged asphalt, finding that the m gradually increased while the stiffness (S) gradually decreased, which was favorable to the low-temperature cracking resistance of the rejuvenated asphalt. Gong et al. [57] reported that biodiesel-rejuvenated asphalt exhibited better low-temperature flexibility than aged asphalt, thereby reducing the risk of cracking. Zeng et al. [139] found that adding 20% CO derivatives could restore both the m and S of rejuvenated asphalt compared to a virgin sample, thereby improving the low-temperature cracking resistance of rejuvenated asphalt.

In addition, Hong et al. [23] found that a compound rejuvenator consisting of AO and SBS not only prevented the excessive softening effect on aged asphalt caused by AO but also enhanced its low-temperature properties. This combination could mitigate AO's impact on the rutting resistance of rejuvenated asphalt. Similarly, a compound rejuvenator composed of WEO and SBS was found to restore aged asphalt's low-temperature cracking resistance [79]. As shown in Figure 16, modified alkyl amino polyamines could cause a decline in the S of rejuvenated SBSMA, although S remained higher than virgin sample. The phenomenon suggested that the rejuvenator was able to effectively restore the low-temperature properties of aged SBSMA [140]. Furthermore, a compound rejuvenator consisting of TMPGE and ESO reduced the S value and increased the m of rejuvenated SBSMA. This made it clear that it could restore the cracking resistance at low temperatures to that of a virgin sample [122].

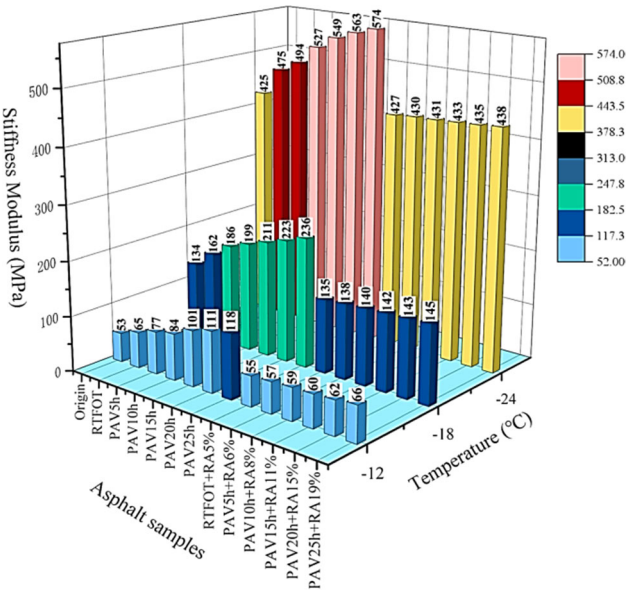


Figure 16. Diagram of the change in m in rejuvenation to different degrees of aging SBSMA (Pressure Aging Vessel (PAV) and Rolling Thin Film Oven Test (RTFOT)) [140].

5. Macro-Properties of Rejuvenated Asphalt Mixture

5.1. High-Temperature Stability

The rutting resistance is typically applied to assess the high-temperature stability of asphalt mixture [141]. Zaumanis et al. [142] concluded that aromatic extracts had less impact on the rutting depth of asphalt mixture compared to WEO, attributing this to the fact that low-polar molecules in WEO further softened the rejuvenated asphalt. Based on this, Chen et al. [70] researched variations in the properties of WEO-rejuvenated asphalt mixture using dynamic uniaxial compression tests and Hamburg Wheel Tracking Tests. The results showed that the rutting resistance of an WEO-rejuvenated asphalt mixture was superior to that of a virgin sample. Wen et al. [143] evaluated how the WCO worked on RAP and analyzed the changes in high-temperature stability of rejuvenated asphalt mixtures using dynamic creep tests. The experimental results showed that the rutting resistance of an asphalt mixture declined with a high dosage of WCO. Hohmann et al. [144] found that higher epoxy values in soybean compound oil led to increased rutting depth in rejuvenated asphalt mixtures, suggesting that higher epoxy values reduced rutting resistance. As shown in Figure 17, among the six rejuvenators added to the 100% RAP mixture, the mineral oil-based rejuvenators recovered rutting depth better than the bio-based rejuvenators [142].

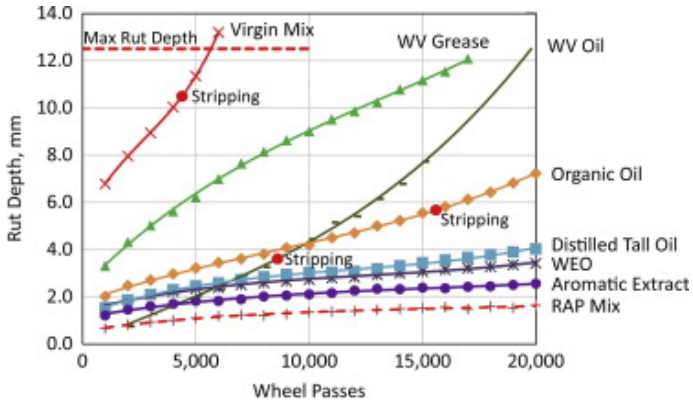


Figure 17. Hamburg Wheel Tracking Tests rutting results [142].

In addition, Eltwati et al. [79] found that a compound rejuvenator consisting of WEO and SBS was able to cause the flow value of a rejuvenated asphalt mixture to rise and improve its rutting resistance. This performance was further improved as the rejuvenator content increased. Chen et al. [119] found that higher-epoxy-value compounds enhanced reactions with sulfur–oxygen compounds in a rejuvenated asphalt mixture, making it easier to form a three-dimensional network structure, which improved the rutting resistance and elasticity of the rejuvenated asphalt mixture. Interestingly, Cao et al. [145] investigated how the HDI and MDI affected rutting resistance of aged SBSMA mixtures. They discovered that their addition caused the rutting resistance to degrade, as indicated by rutting tests. Furthermore, Daryaee et al. [146] found that incorporating waste polymer and soft asphalt increased the flow value of the mixture and upgraded the high-temperature stability of RAP.

5.2. Fatigue Resistance

Fatigue resistance is often applied to evaluate the medium-temperature stability of asphalt mixtures [147]. Zaumanis et al. [142] analyzed six rejuvenators to assess the performance of asphalt mixture containing 100% RAP. It was clear that RAP rejuvenated by WEO exhibited the fewest failure cycles and was more prone to fatigue cracking, as determined by the co-axial shear test. Furthermore, Gong et al. [148] conducted a four-point flexural fatigue life test and found that BO could recover the fatigue cracking resistance of RAP-containing asphalt mixture to that of a virgin sample. Additionally, Daryaee et al. [146]

assessed how AO and WPE compound rejuvenators worked on the fatigue resistance in asphalt mixture containing 50% RAP. Their findings showed that the stiffness of the mixture decreased when adding rejuvenators, thereby enhancing its fatigue resistance. Moreover, Cao et al. [145] researched how HDI and MDI affected the fatigue resistance of an aged asphalt mixture through beam bending experiments. They discovered that the fatigue life increased to 145,120 cycles when reactive rejuvenators were added, significantly reducing the potential for fatigue failure at the mixture interface and effectively restoring its fatigue resistance. Shu et al. [149] found that adding AO to PU repaired the broken SBS chains and supplemented the missing light components in rejuvenated asphalt mixtures, reducing the possibility of fatigue cracking in rejuvenated asphalt mixtures.

Low-Temperature Cracking Resistance

The cracking resistance of asphalt mixtures at low temperatures is an important index to reflect their performance. Zhang et al. [150] investigated how WCO affected the performance of an asphalt mixture containing 60% RAP. They found that the WCO-rejuvenated asphalt mixture's maximum bending strain was higher than specification value through three-point bending tests, which indicated that the mixture could better recover its low-temperature resistance. Additionally, Zaumanis et al. [142] compared the tensile strength of waste vegetable oil (WVO)- and organic-oil-rejuvenated asphalt mixture. They concluded that WVO more effectively restored low-temperature crack resistance compared to organic oil.

Furthermore, Daryaee et al. [151] investigated how a compound rejuvenator consisting of polymers and AO affected an asphalt mixture containing 50% RAP. They came to the conclusion that the rejuvenator upgraded the cracking resistance of the mixture at low temperatures by softening the asphalt. Cao et al. [145] researched how HDI and MDI affected the cracking resistance of an aged SBSMA mixture at low temperatures. The results showed that the fracture strain and flexural strength increased when rejuvenators were added. Hu et al. [101] compared the difference between the carrier method (new asphalt directly mixed with rejuvenators) and the non-carrier method on the performance of PU- and BUDGE-compound-rejuvenated asphalt mixture. Through beam bending tests, they found that compared to the non-carrier method, the carrier method could better recover low-temperature flexural strength of the rejuvenated asphalt mixture. This caused a more obvious improvement regarding the cracking resistance of mixture at low temperatures. In summary, various kinds of rejuvenators produce varying impacts on the cracking resistance of rejuvenated asphalt mixtures at low temperatures. One should be cautious to alter rejuvenators based on the specific requirements of the situation.

5.3. Water Stability

Early deterioration of pavement is mainly caused by water damage, making water stability a significant performance feature of asphalt mixture. Therefore, ensuring a rejuvenated asphalt mixture's water stability is essential for maintaining road durability. Chen et al. [70] evaluated how the WEO affected the water stability of rejuvenated asphalt mixtures and drew a conclusion that asphalt mixtures containing 50% RAP rejuvenated with WEO exhibited good water stability. Similarly, Zaumanis et al. [142] investigated the water stability of WVO- and organic-oil-rejuvenated asphalt mixtures. Their results from Hamburg Wheel Tracking Tests showed that WVO exhibited an inflection point, whereas organic oil did not, indicating that WVO-rejuvenated asphalt mixture had poor water stability and did not meet the required performance standards. Additionally, Ahmed et al. [9] found that free fatty acids, which are hydrophilic components presenting in untreated

WCO, negatively impacted the water stability of a rejuvenated asphalt mixture, further compromising its durability.

Conversely, Eltwati et al. [80] found that the Tensile Strength Ratio (TSR) value decreased with increasing RAP content. However, AO and SBS demonstrated a strong hydrophobicity, effectively restoring the water stability of the rejuvenated asphalt mixture to that of a virgin sample. Similarly, Cao et al. [145] researched how HDI and MDI affected the water stability of a rejuvenated asphalt mixture using freeze–thaw splitting experiments. Their outcomes showed that the freeze–thaw split strength ratio was recovered to that of a virgin sample after the addition of rejuvenators. Furthermore, Han et al. [94] examined how BUDGE and PU affected the water stability of a rejuvenated asphalt mixture. Their study showed that the rejuvenators effectively enhanced the rejuvenated asphalt mixture's water stability.

6. Technical Challenges and Future Recommendations

As one of the main materials for road pavement, the demand for asphalt mixtures continues to increase. However, the increasing service life of pavement produces a large amount of RAP, which not only causes a waste of resources, but also causes potential pollution to the environment. Therefore, how to effectively utilize RAP and realize environmental protections has become a hot topic. The use of rejuvenators is one of the key technologies to restore the properties of aged asphalt. Based on the existing literature, this review summarizes the current status of the application of rejuvenators in road engineering, then reminds readers of current problems and finally looks forward to future development trends.

In actual projects, the classification and standardization of rejuvenators is an urgent problem. Based on differences in composition, this paper classifies rejuvenators into mineral oil-based, bio-based, and compound rejuvenators. This categorization method works best for the distinction between mineral oil-based and bio-based rejuvenators. However, it is difficult to subdivide the compound rejuvenators because the compound rejuvenators have a complex composition and still lack a unified division standard. Based on the current situation, for the sake of further improving the classification system of rejuvenators, it is essential to establish a unified standard for the division of compound rejuvenators to better guide the selection and application of rejuvenators.

At present, the classification of rejuvenators is mainly based on two aspects: composition and property. In initial past research, researchers mainly focused on the rejuvenation of aged asphalt, and usually oil substances similar to the composition of asphalt were added as rejuvenators, which restored low-temperature resistance but sacrificed high-temperature resistance. Currently, mineral oil-based and bio-based rejuvenators are representative choices. Although the use of fresh oil is more common, waste oil has also been gradually applied during recent years to recover the properties of aged asphalt. Specifically, some fresh oil may still have a high potential for reuse after daily use. Through proper treatment and modification, waste oil can recover the properties of aged asphalt to a certain extent. For example, waste motor oil, waste cooking oil, etc., can be processed by distillation, filtration, and centrifugal separation to obtain rejuvenators with a similar composition to asphalt. This not only realizes the recycling of resources but also prevents waste oil from polluting the environment. Therefore, the reuse of waste oil is a future research direction.

In addition, in order to realize more efficient rejuvenation, the research and development of compound rejuvenators is especially important. In previous research, compound rejuvenators were mainly designed for aged modified asphalt through the addition of new modifiers or in situ reconstruction of degradation of modifiers to better rejuvenate aged modified asphalt [16]. The direct addition of modifiers was able to shape a new

three-dimensional network structure, which was capable of comprehensively improving the properties of rejuvenated modified asphalt. Although this rejuvenation method is simple and effective, it does not allow for degraded modifiers to be recovered, causing an inefficient utilization of resources. In contrast, the in situ reconstruction method reconstructs the network structure in situ through the reaction between the reactive rejuvenators and groups in the degraded modifier. It can simultaneously upgrade the high- and low-temperature resistance of rejuvenated modified asphalt. Therefore, in situ reconstruction of degraded modifiers is still a key point of interest in upcoming studies. According to the previously mentioned studies, it can be found that the isocyanate system and epoxy systems are the main research directions at present, and these discoveries offer guidance for further studies on reactive rejuvenators. However, reactive rejuvenators are highly toxic and harmful to the environment and human health. On this basis, some scholars have begun to modify isocyanate-type regenerators, such as Han et al. [152], who modified isocyanates with chain expanders and found that, compared with traditional rejuvenators, the effect of the modified rejuvenators is more significant, and the toxicity of isocyanates is greatly reduced. Therefore, chemical modification of existing rejuvenated agents will be the main research direction in the future. However, this method still has the disadvantages of high cost and cumbersome synthesis steps. In addition, the concept of single-expansion regenerators has also emerged, which restores the molecular weight of broken SBS by unidirectionally replenishing chain segments, thereby restoring the properties of aged asphalt. Nevertheless, present studies still have obvious disadvantages. On the one hand, available studies are mainly interested in the research and development of two types of compounds, epoxy-system and isocyanate-system compounds, ignoring reactions between other active compounds and degradation modifiers. In addition, the labor and material costs of the research process are high. In fact, according to Yan et al. [153], computer neural networks were used to simulate how different groups react with aged asphalt, which could assess the rejuvenation potential of active reactants. Using neural networks reduces the experimental time and improves the efficiency of research and development at the same time. It provides new ideas for the intelligent design of rejuvenators. In addition, through molecular dynamics and quantum chemistry techniques, the mechanism of action of rejuvenators can be understood more deeply, and the design of rejuvenators can be further optimized. Further, although the micro- and macro-properties of recycled asphalt have been studied in depth, the relationship between the two is unclear. Researchers use changes in macroscopic properties to explain the changes in the microscopic properties of asphalt, but they cannot establish the relationship between the two through mathematical models. Therefore, a direction of future research is to establish mathematical models of variations between micro and macro performance.

Finally, the property evaluation of rejuvenated asphalt is the key to ensuring its stable properties in practical applications. Currently, instruments such as AFM, FM, and SEM are often used to analyze the microstructure of asphalt, but these methods are still lacking in the study of the morphology of asphalt rejuvenated by compound rejuvenators. For the macroscopic property assessment of rejuvenated asphalt mixtures, current methods and standards are not uniform, which leads to difficulties in the quality control and engineering application of rejuvenated asphalt mixtures. In addition, the excessive use of some rejuvenators may, on the contrary, negatively affect the properties of asphalt, and these limitations can lead to the impossibility of accurately assessing the rejuvenation efficiency of rejuvenators. Therefore, improving microscopic evaluation methods of compound rejuvenators and accurately controlling the number of rejuvenators are effective ways to improve the rejuvenation efficiency of rejuvenators.

7. Conclusions

Rejuvenation of aged asphalt has important economic and environmental advantages. The current research status of asphalt rejuvenators was reviewed systematically in this paper. In this paper, the recovery effect between different rejuvenators was systematically evaluated. At the same time, the current rejuvenator classification system and micro- and macro-properties were analyzed in depth, which filled some gaps in the research field of rejuvenators. Based on this, areas of interest for future research were discussed and suggested. Regarding the review results, the following conclusions were obtained:

1. Rejuvenators can be classified into mineral oil-based rejuvenators, bio-based rejuvenators, and compound rejuvenators according to differences in composition. Among these rejuvenators, mineral oil-based and bio-based rejuvenators are the most researched ones, and compound rejuvenators are the most promising ones for application.
2. Through micro-experiments, it can be found that mineral oil-based and bio-based rejuvenators, through physical dilution of aged virgin asphalt to upgrade its microscopic performance, can only restore the smoothness of aged asphalt without changing the microstructure of aged asphalt. Oil-based and polymer compound and reactive rejuvenators can not only restore the properties of asphalt through physical dilution but also reconstruct the original structure or form new structures by supplementing degraded modifiers and repairing broken modifiers. This can restore the properties of aging modified asphalt.
3. Mineral oil-based and bio-based rejuvenators are able to obviously enhance the low-temperature resistance of aged asphalt, although the high-temperature resistance is reduced by the presence of low-polarity molecules and excessive softening of the asphaltenes. Reactive rejuvenators can reconnect degraded SBS modifiers and rejuvenate aged virgin asphalt, which can recover the performance of aged SBSMA, even to the level of virgin sample. In addition, bio-based and mineral oil-based rejuvenators can also be modified to mitigate the loss of high-temperature resistance caused by excessive softening.
4. Untreated waste bio-oil (excessive free fatty acids) and waste mineral oil (containing metal ions) rejuvenators can adversely affect the water stability of asphalt mixtures. In addition, compound rejuvenators are significantly better than bio-based and mineral oil-based rejuvenators in restoring high-temperature resistance and water stability to aged asphalt mixtures.
5. Currently, this technology has not been applied widely to the field of engineering. In the future, it is essential to consider artificial intelligence combined with rejuvenator design and modify waste oil or modify current reactive rejuvenators to better rejuvenate aged asphalt.

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Abbreviations

RAP	Reclaimed Asphalt Pavement
SBS	Styrene-Butadiene-Styrene
SBSMA	SBS Modified Asphalt
AO	Aromatic Oil
WEO	Waste Engine Oil
WCO	Waste Cooking Oil
GO	Gutter Oil
SO	Soybean Oil
CO	Castor Oil
TO	Tung Oil
BO	Bio Oil
MO	Motor Oil
PU	Polyurethane
TAIC	Triallyl Isocyanate
MDI	4,4-Diphenyl-ethane-diisocyanate
TDI	2,4-Toluylene Diisocyanate
TGDOM	4,4-Diphenyl-methane-diisocyanate
ESO	Epoxidized Soybean Oil
BUDGE	1,4-Butanediol Di glycidyl Ether
TMPGE	Trimethylolpropane Tri glycidyl Ether
HDDGE	1,6-Hexanediol Di glycidyl Ether
BDMA	N,N-Dimethylbenzylamine
RDGE	2,2′-[1,3-Phenylenebis Oxyethylene)] Dioxirane
PEHA	3,6,9,12-Tetraazatetradecane-1,14-diamine
TPP	Triphenylphosphine
DSR	Dynamic Shear Rheometer

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